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**6. AUTHOR(S)**
P.I. Cohen

**7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(ES)**
Department of Electrical Engineering
University of Minnesota
200 Union St. SE
Minneapolis, MN 55455

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FINAL TECHNICAL REPORT

# Epitaxial Metallizations for III-V Semiconductors

AF/F9620-94-1-0330

Submitted by

P.I. Cohen
Department of Electrical Engineering
University of Minnesota
200 Union St. SE
Minneapolis, MN 55455

## Abstract

The nucleation and growth of three intermetallic systems on III-V semiconductors have been examined or begun. FeAl and CoGa have been grown on GaAs(100) and their structural and magnetic properties examined. For their nucleation and growth, the main techniques used were ultrahigh vacuum scanning tunneling microscopy and reflection high energy electron diffraction. Direct confirmation of the growth transition from single-layer to bilayer for FeAl was obtained. Magneto-optical Kerr effect measurements showed the transition in magnetic properties of FeAl. These results suggest that fundamental aspects crucial to spin-valve devices could be studied without the complication of a heterointerface. Studies of CoGa on GaAs showed how to prepare smooth films of CoGa and observed a fourfold anisotropy. However it left unanswered the controversy of whether lattice matching would allow preparation of pinhole free films. Finally, an effort to grow prepare high quality GaN was started, to serve as a substrate for the *in situ* growth of HfN films. A source was constructed and GaN films were prepared. However, deposition of this promising material has not yet been attempted.

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# 1 FeAl Nucleation and Growth on GaAs(100)

Brian Ishaug was supported on this grant to study the growth of FeAl on GaAs. He used scanning tunneling microscopy and reflection high energy electron diffraction to examine the nucleation and growth of the FeAl film. This is an interesting system since the FeAl undergoes a magnetic transition and an ordering transition at a composition of Fe:Al of 3:1. The structure of FeAl is CsCl in which the (100) planes alternate between Fe and Al. For $Fe_3Al$, the Al plane becomes a checkerboard of Fe and Al. This bulk superlattice was expected to have different growth kinetics and different magnetic properties.

An investigation of the nucleation and growth as well as magnetic properties of epitaxial $Fe_xAl_{1-x}$ on AlAs/GaAs(100) was conducted. In-situ reflection high-energy electron diffraction (RHEED) and ultrahigh vacuum (UHV) scanning tunneling microscopy (STM) were used to characterize the surface and ex-situ magneto-optical Kerr effect (MOKE) measurements were used to characterize the magnetic properties. It was found that epitaxial films can be grown over a broad composition range $0.5 < x < 0.8$ provided the appropriate nucleation procedure is used, most important of which is the deposition of more than 90Å of $Fe_xAl_{1-x}$ before annealing. $Fe_xAl_{1-x}$ undergoes an unusual incubation effect over the first 3 bilayers of growth on AlAs. STM images taken at 1 and 3 bilayers shed some light on why this incubation effect exists. After depositing 90Å and annealing, the films exhibited a $(2 \times 2)$ and/or a $(5 \times 5)$ surface reconstruction. This reconstruction depended upon the anneal temperature and film composition. STM images of a typical annealed film showed atomic step terraces with step heights roughly corresponding to the height of an $Fe_xAl_{1-x}$ bilayer. Differences were also seen in the surface morphology of the 2-fold and 5-fold surfaces. Growth of $Fe_xAl_{1-x}$ on an annealed $Fe_xAl_{1-x}$ surface produced RHEED oscillations which were found to occur in 2 distinct modes, monolayer and bilayer. It was determined that this growth mode was primarily dependent upon growth composition, but had some dependence on the annealed $Fe_xAl_{1-x}$ starting surface. The composition at which the growth mode changed from monolayer to bilayer closely corresponded to the composition at which $Fe_xAl_{1-x}$ changes from ferromagnetic to nonmagnetic. Magnetic measurements of several samples confirmed samples above $x = 0.7$ to be ferromagnetic with magnetization in-plane. A compositional dependence on coercivity and saturation magnetization was also found.

Brian Ishaug presented the results of this work at a late news paper at the National Symposium of the American Vacuum Society in October, 1995 and a more complete version at the National meeting of the Materials Research Society in Boston, in November, 1995. After more measurements were made, including the magnetic measurements described above the results were included in a M.S.E.E. thesis and published in Surface Science.

## 2 CoGa ON GaAs(100)

We examined the microscopic processes involved in the nucleation and growth of epitaxial CoGa on GaAs. In particular we the morphological stability of epitaxial CoGa films using in situ RHEED and UHV-STM as well as ex situ RBS was studied. Films were grown by codeposition of Co and Ga directly on a GaAs surface at several deposition temperatures and monitored by measuring RHEED intensity oscillations. Films were then annealed to between 250 and 580°C. We found that the surface morphology varied significantly with growth and anneal temperature. Growth of CoGa at 360 C and annealing to 520°C results in the formation of a network of coalesced islands, still showing a remnant of their square anisotropy. For a 15 nm deposition, the coalesced islands are typically 150 nm wide and as much as 1.3 $\mu$m long, covering about 60% of the surface. By contrast, growth of CoGa at 150 C and annealing to 250 and 550°C resulted in much smoother films. For the 250 C anneal, the surface exhibited a $2 \times 2$ RHEED pattern and STM showed small interconnected grains. The individual grains were about 4 nm $\times$ 4 nm in size and 1 bilayer high. For the 550°C anneal, the surface exhibited a $(4 \times 4)$ RHEED pattern. STM exhibited large atomically smooth terraces, 100 to 200 nm in size with atomic corrugation of 8.1 Å, characteristic of a 4-fold reconstruction of CoGa. In comparison CoAl grown at 150°C and annealed to 580 C produced similar results to the CoGa film annealed to 550°C but with pin holes as large as 120 nm and 6 nm deep covering about 20% of the surface. This is the first combined diffraction and atomic-resolution STM study of the nucleation and growth of atomically smooth, thermodynamically stable intermetallic films. These results were presented at the National Symposium of the Materials Research Society in Boston, December, 1996.

## 3 Growth of HfN on GaN$(000\bar{1})$

HfN should be a thermodynamically stable analogue of these same intermetallic structures on GaN. It is nearly lattice matched and we predict would provide an abrupt Ohmic contact. To examine this part of the funds provided by this AASERT were used to support Sean Seutter and Jason Borton to initiate work on the growth of GaN. This was a crucial first step to determine the nature of the contact. Without high quality GaN, no electrical characterization could be done. Without high quality GaN no *in situ* structural analysis could be performed. These two students were instrumental in determining the GaN growth conditions (Seutter) and etching procedures (Borton) to be able to examine HfN/GaN contacts. We set up a Varian Gen II MBE for the growth of GaN using $NH_3$ and Ga. GaN has been growth successfully, with advances made in surface characterization and in the nucleation and growth of single domain GaN. This is ongoing work, currently supported by the

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# Nucleation, growth and magnetic properties of epitaxial FeAl films on AlAs/GaAs

B.E. Ishaug [a], S.M. Seutter [a], A.M. Dabiran [a], P.I. Cohen [a,*], R.F.C. Farrow [b], D. Weller [b], S.S.P. Parkin [b]

[a] Department of Electrical Engineering, University of Minnesota, Minneapolis, MN 55455, USA
[b] IBM Almaden Research Center, San Jose, CA 95120, USA

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B.E. Ishaug [a], S.M. Seutter [a], A.M. Dabiran [a], P.I. Cohen [a,*], R.F.C. Farrow [b], D. Weller [b], S.S.P. Parkin [b]

[a] *Department of Electrical Engineering, University of Minnesota, Minneapolis, MN 55455, USA*
[b] *IBM Almaden Research Center, San Jose, CA 95120, USA*

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**Abstract**

An investigation of the nucleation and growth as well as magnetic properties of epitaxial $Fe_xAl_{1-x}$ on AlAs/GaAs(100) is reported. In situ RHEED and UHV STM were used to characterize the surface and ex situ MOKE measurements were used to characterize the magnetic properties. We found that epitaxial films can be grown over a broad composition range, $0.5<x<0.8$, provided the appropriate nucleation procedure is used, most important of which is the deposition of more than 90 Å of $Fe_xAl_{1-x}$ before annealing. STM images taken at 1 and 3 bilayers of growth reveal a surface covered in small islands 40 Å in size and 2 Å high. After depositing 90 Å and annealing, STM images reveal a much smoother surface with atomically flat terraces greater than 100 Å in size. STM images also showed step heights corresponding to the height of an $Fe_xAl_{1-x}$ bilayer. In addition, the annealed films exhibited a $(2\times2)$ and/or a $(5\times5)$ surface reconstruction as determined by RHEED. The $(2\times2)$ reconstruction was seen after annealing the films to 550°C and the $(5\times5)$ reconstruction would begin to show up after annealing films of composition $x>0.55$ to 650–700°C. STM images of the $(5\times5)$ surface revealed a much lower step edge kink density than the $(2\times2)$ surface. When growing $Fe_xAl_{1-x}$ on an annealed $Fe_xAl_{1-x}$ surface, RHEED oscillations were found to occur in two distinct modes, monolayer and bilayer, where monolayer growth would occur at $x>0.7$ and bilayer growth at $x<0.7$. Excess Fe on the annealed surface, such as what is encountered on a $(5\times5)$ surface, could force a bilayer growth mode to monolayer growth mode for several layers of growth. This was due to the excess Fe getting incorporated into the growth front. Magnetic measurements showed samples above $x=0.7$ to be ferromagnetic with magnetization in-plane. A compositional dependence on coercivity and saturation magnetization was also found where higher Fe concentrations corresponded to higher coercivities and higher saturation magnetizations.

*Keywords:* Growth; Magnetic phenomena; Metal–semiconductor interfaces; Nucleation

## 1. Introduction

The growth of stable epitaxial metals on III–Vs is important to metal/semiconductor devices that need to operate at high temperature and to buried metal-layer devices where interdiffusion at interfaces cannot be permitted. However, most metals that can be grown epitaxially on III–Vs are not thermodynamically stable so that even modest heating causes reaction and interdiffusion at the interface. Sands et al. [1] have reviewed these issues and discussed other criteria necessary for stable epitaxial growth of metals on III–Vs. The main conclusion is that binary intermetallics that are lattice matched to III–Vs and which can coexist with the III–V according to bulk phase diagram

*Corresponding author. Fax: +1 612 625 3300.


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are candidate materials. Experiment has confirmed many of these ideas [1], but the interplay of kinetics and thermodynamics as well as the details of the growth mechanisms have not been worked out. The main purpose of this paper is to combine scanning tunneling microscopy (STM) with reflection high-energy electron diffraction (RHEED) to study these competing issues.

Two major groups of intermetallics have been found to form stable epitaxial films. The first group has a cubic CsCl (B2) crystal structure, known collectively as transition metal-three (TM-III) intermetallics. Some notable examples include NiAl, CoAl, FeAl, NiGa and CoGa. Their lattice parameters are usually 1–3% larger than one half the lattice parameter of GaAs and their CsCl phases usually exists over a wide composition range. The second major group has a NaCl (B1) structure and is known collectively as rare earth-five (RE-V) intermetallics. Some notable examples include ErAs, LuAs and (Sc,Er)As. Their lattice mismatch is generally smaller, less than 2%.

In this paper we examine the $Fe_xAl_{1-x}$ system because, in addition to the CsCl phase, it has a stable $BiF_3$ ($DO_3$) phase that is also reasonably well lattice matched to GaAs. The CsCl phase exists over the composition range $0.5<x<0.63$ and has a lattice mismatch of 2.9% while the $BiF_3$ phase exists over the composition range, $0.63<x<0.78$ [2] and has a lattice mismatch of 2.4% [3]. Both phases have been shown to form stable epitaxial films on AlAs up to 550°C [4,5] and the $BiF_3$ phase has even been successfully grown directly on GaAs [6,7]. In addition, $Fe_xAl_{1-x}$ is particularly interesting since it becomes ferromagnetic at compositions above $x=0.7$. This makes it possible for $Fe_xAl_{1-x}$ to be used for magnetic metal/semiconductor devices. Further, its magnetic properties can be varied in a given structure simply by varying its composition.

In this paper we investigate nucleation, annealing, growth and magnetic properties of $Fe_xAl_{1-x}$ films spanning both the CsCl and $BiF_3$ composition range, $0.5<x<0.78$. Previous work by Kuznia, Wowchak and Cohen used RHEED and Auger electron spectroscopy to study the nucleation and growth of $Fe_xAl_{1-x}$ on pseudomorphic AlAs on InP(100) [4] and on AlAs/GaAs(100) [5]. They reported several main results. First, the initial nucleation exhibits an incubation period lasting for 4 to 6 monolayers of growth, in which the intensity of the diffraction pattern decreases to near zero and then recovers (see Fig. 2). Second, growth of $Fe_xAl_{1-x}$ on an annealed $Fe_xAl_{1-x}$ surface results in RHEED oscillations with two distinct oscillation frequencies, one corresponding to monolayer-by-monolayer growth and one corresponding to bilayer-by-bilayer growth where a bilayer is defined as a two atom thick layer and a monolayer is defined as a one atom thick layer (Fig. 1). These previous studies had difficulty extracting quantitative information on the surface morphology from RHEED measurements, especially in the nucleation stage where the diffraction was weak and diffuse. In this work, we applied UHV STM to obtain quantitative information on the real space surface, including step heights, terrace lengths and island sizes. In addition, we studied higher anneal temperatures and growth of films with higher Fe composition in more detail. Finally, we examined the magnetic properties of these films to see if a thin film behaves the same as bulk material.

## 2. Experimental

A PHI-400 solid source MBE system was used for growth. Standard Knudsen cells were used for all sources except Fe, for which an electron bombardment source similar to that described by Jonker et al. was used [8]. The system was also equipped with a quartz crystal deposition monitor which could be positioned directly in front of the substrate. This was used to measure the Fe and Al

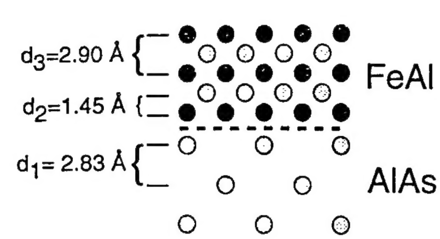


Fig. 1. Diagram showing an AlAs monolayer – $d_1$, an FeA monolayer – $d_2$, and an FeAl bilayer – $d_3$.

flux prior to growth. In situ growth monitoring was performed with a 10 keV RHEED system on the growth chamber and the STM was housed in a separate chamber attached to the growth chamber. Samples could be transferred to this chamber without exposure to pressures above $1 \times 10^{-8}$ Torr. MOKE measurements were performed in air using a laser with a wavelength of 650 nm.

Epi-ready GaAs(100) $n^+$Si doped samples were cleaned by desorbing the oxide at 620°C in an $As_4$ background of $1 \times 10^{-6}$ Torr. A 3000 Å GaAs buffer, doped with Si to about $1 \times 10^{18}$ atoms $cm^{-3}$, was then grown. The buffer and substrate were doped to ensure that the sample would be conducting for STM measurements. After the GaAs buffer, 10 ML of AlAs was deposited at 540°C. With AlAs as an effective substrate, the thermodynamic stability argument regarding a three-element system is expected to apply [1]. The sample was cooled to 200°C and removed from the deposition chamber. The As source was then cooled and the remaining As background was gettered with Ga and Al until the base pressure was below $3 \times 10^{-9}$ Torr. The Fe and Al sources were then set to give the desired Fe–Al ratio using the quartz crystal deposition rate monitor. The sample was then returned to the deposition chamber and heated to 700°C until an AlAs(3×2) pattern appeared. This procedure was shown to drive-off Ga surface contaminates and reduce the As coverage on the surface [9]. Finally, the sample temperature was brought back down to 200°C and $Fe_xAl_{1-x}$ was grown by co-deposition of Fe and Al.

To produce an epitaxial film, we deposited a minimum of 90 Å of $Fe_xAl_{1-x}$ on the AlAs surface before annealing to between 550 and 700°C for 5 min. After annealing, additional $Fe_xAl_{1-x}$ was grown by co-deposition at 200°C, during which time RHEED oscillations were obtained. The RHEED oscillations would die out after growing about 100 Å. Shortly after oscillations died out, the film would again be annealed to between 550 and 700°C for 5 min. This process of growth and annealing could be repeated indefinitely without degradation of the RHEED pattern or RHEED oscillations. Using the RHEED oscillations and the assumption that the Al source flux stayed constant, the composition of the $Fe_xAl_{1-x}$ film could be determined at any time during growth. This composition ratio was used as a verification of the ratio determined by the quartz crystal deposition monitor, and the two were generally within 5% of each other. The RHEED oscillation value was also used to adjust for drift in the Fe flux, which is known to have a significant drift problem due to the e-beam heating method. In addition, because oscillations are expected to be a more accurate determinant of composition, the composition values reported in this paper were all determined by RHEED oscillations.

## 3. Results and discussion

### 3.1. Nucleation

In Fig. 2, the intensity of the specular diffracted beam is shown versus time during the initial growth of FeAl on an AlAs buffer. Here, $Fe_xAl_{1-x}$ is co-deposited at a rate of 0.25 monolayers (ML) per second, which corresponds to a period of 4 seconds per monolayer. The nucleation of the film is characterized by a near complete drop in the diffracted intensity followed shortly thereafter by a recovery and, in some cases, weak intensity

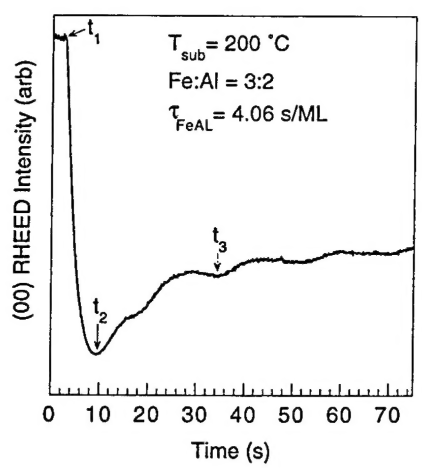


Fig. 2. RHEED intensity of the specular diffraction beam during initial nucleation of $Fe_xAl_{1-x}$ on AlAs. The starting pattern at $t_1$ is an AlAs(3×2) pattern. Growth is started at $t_1$. At time $t_2$, the RHEED pattern is no longer visible. At time $t_3$, the intensity recovers to 30% of the original intensity and exhibits a broad and diffused (1×1) pattern.

oscillations. The starting RHEED pattern at $t_1$ is an AlAs(3×2). At $t_2$, after about 1–2 ML of deposition, the pattern is gone and the RHEED exhibits only a diffuse background. With continued deposition a weak (1×1)$Fe_xAl_{1-x}$ pattern begins to develop. At $t_3$, after 6 ML of deposition, the (1×1) pattern is now between 30 and 50% of the original intensity and remains near this intensity with continued deposition. The time period over which the diffraction pattern remains near zero is termed the incubation period and the time between when the pattern begins to recover and is fully recovered is termed the recovery period.

Figs. 3a and 3b shows STM images taken after roughly 2 ML of growth. This corresponds to the point in the nucleation where the RHEED pattern intensity is a minimum. The first image, Fig. 3a, shows a long-range scan. Large terraces, some greater than 120 nm in size, can be seen in this image. A close range scan, Fig. 3b, shows that the entire surface is covered with small features, 40 Å in size and approximately 2 Å high. These small features do not appear to be clearly resolved and are likely a convolution of the real features on the surface and the STM tip. Because of this, the real height values may be somewhat larger than the measured values. We interpret the large terraces observed in Fig. 3a to be due to the underlying AlAs surface and the small features due to $Fe_xAl_{1-x}$ clusters. Noting that there is only diffuse scattering in the RHEED pattern, we conclude that the small clusters are either disordered or randomly oriented on the AlAs surface.

### 3.2. Annealing

At least 90 Å of $Fe_xAl_{1-x}$ was deposited on the AlAs surface at 200°C before annealing. Attempting to anneal films less than 90 Å thick typically resulted in the inability to obtain RHEED oscillations during $Fe_xAl_{1-x}$ overgrowth. We found some evidence that the minimum thickness which can be annealed and still obtain RHEED oscillations during overgrowth was dependent on film composition. At lower Fe compositions, closer to $x=0.5$, we were able to anneal films as thin as 20 Å with satisfactory overgrowth results.

With $Fe_xAl_{1-x}$ growth on an annealed $Fe_xAl_{1-x}$ surface, RHEED oscillations died out after approximately 100 Å of growth at temperatures between 150 and 200°C. At this point, the film exhibited a broad and diffuse (1×1) diffraction pattern. An anneal was necessary to smooth the surface and make it possible to obtain RHEED oscillations with continued $Fe_xAl_{1-x}$ growth. We annealed to between 550 and 700°C and, depending upon film composition and anneal temperature, the film then exhibited either a sharp (2×2) and/or a sharp

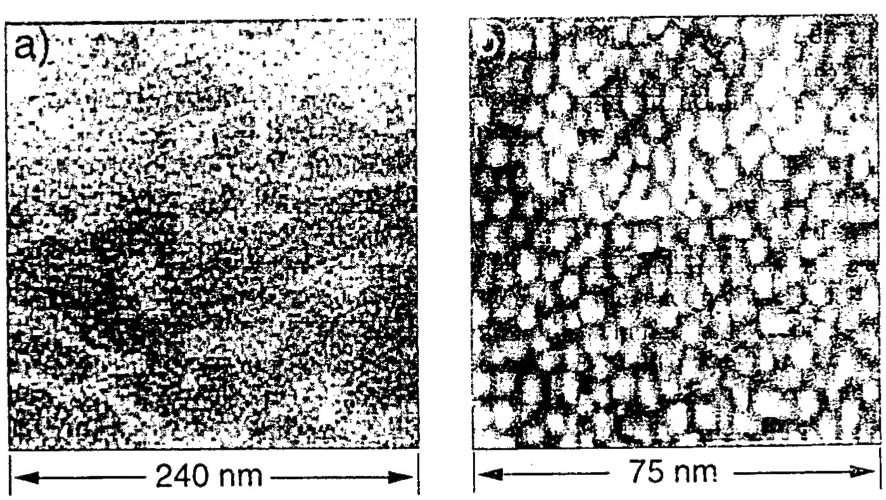


Fig. 3. STM images taken after 2 ML of growth. Terraces from the underlying AlAs surface can be seen in (a) and small clusters of $Fe_xAl_{1-x}$ can be seen in (b).

(5 × 5) diffraction pattern. A sharp (2 × 2) pattern would appear after annealing to 550°C for 5 min. The appearance of this pattern was not dependent on composition. However, annealing films to 650–700°C resulted in the composition dependent formation of a (2 × 2) and/or a (5 × 5) diffraction pattern. At $x=0.5$, the film would exhibit a pure (2 × 2) pattern. Between $0.55<x<0.7$ the film would exhibit a combined (2 × 2) and (5 × 5) pattern with the 5-fold pattern becoming more and more dominant at higher Fe concentrations. At or above $x=0.75$, the film exhibited a pure (5 × 5) pattern. The combined RHEED pattern was a superposition of diffraction patterns from regions of 5-fold ordering and regions of 2-fold ordering as it is impossible for the same surface region to exist with both 2-fold and 5-fold ordering. Although it may appear that the 5-fold ordering is due to the appearance of the $BiF_3$ phase which exists in bulk between $0.63<x<0.78$ at room temperatures, we believe the 5-fold ordering is due to Fe that segregates to the surface during anneal. We came to this conclusion based on results of growing $Fe_xAl_{1-x}$ on an annealed $Fe_xAl_{1-x}$ surface exhibiting a (5 × 5) reconstruction. This is explained in more detail in the growth section.

We compared the surface of two annealed films with UHV STM, one exhibiting a (2 × 2) diffraction pattern and one exhibiting a (5 × 5) pattern. Figs. 4a and 4b show the images corresponding to the (2 × 2) surface. This film had the composition $Fe_{0.72}Al_{0.28}$ and was annealed to approximately 550°C. Islands and terraces are clearly visible. The average island size is about 150 Å and average step height is about 3 Å, which roughly corresponds to one half of the $Fe_3Al$ lattice constant 5.79 Å. Figs. 4c and 4d show STM images corresponding to a (5 × 5) surface. This film had the composition $Fe_{0.76}Al_{0.24}$ and was annealed to approximately 650–700°C. Here, few islands are visible, but terraces can be seen. The average terrace length is about 400 Å and the measured step height is approximately 3 Å. Comparing the two images, it is clear that the (5 × 5) has a much smaller step density than the (2 × 2) surface, as is expected by a higher temperature anneal. The (5 × 5) surface also exhibits a strong crystalline anisotropy along the $(0\bar{1}1)$ and the (011) directions, causing the appearance of sharp 90° kinks in the terrace step edges.

### 3.3. Growth

After annealing, $Fe_xAl_{1-x}$ was deposited at various compositions. Depending on the growth composition and the preparation of the surface, two different growth modes were observed, monolayer-by-monolayer and bilayer-by-bilayer, as determined by the period of the oscillations in the specular RHEED intensity. Compositions of $0.7<x<0.8$ always resulted in monolayer growth while compositions of $0.5<x<0.7$ resulted in either bilayer or a combined monolayer and bilayer growth. In this composition range, the annealed surface upon which we were growing had a large effect on growth mode. On a (2 × 2) surface, growth would be bilayer; however, when attempting to grow on a surface exhibiting a (5 × 5) diffraction pattern, at least the first few layers would grow monolayer-by-monolayer. The duration of the monolayer growth was dependent on the growth composition. For example, at a composition of $x=0.6$ on a (5 × 5) surface, the first 10–12 oscillations would be monolayer oscillations. At a composition of $x=0.5$, only the first one or two oscillations would be monolayer oscillations. The growth mode would then make a transition back to bilayer-by-bilayer growth. Fig. 5 shows RHEED oscillations at various compositions on a (5 × 5) surface. Note that even at $x=0.51$ the first oscillation is a monolayer oscillation and that, as $x$ is increased, the monolayer oscillations persist longer. It can also be seen that the magnitude of the monolayer and bilayer oscillations are different, suggesting separate regions of monolayer growth and bilayer growth. The long-term variation in magnitude and offset of the oscillations are attributed to movement of the diffracted beam into and out of the detection area of the photomultiplier. This beam wandering effect was due to either the changing magnetic properties of the film or the changing electrical transport properties of the sample during growth. We were using a heating method in which electrical current was passed directly through the sample and any changes in the electrical transport proper-

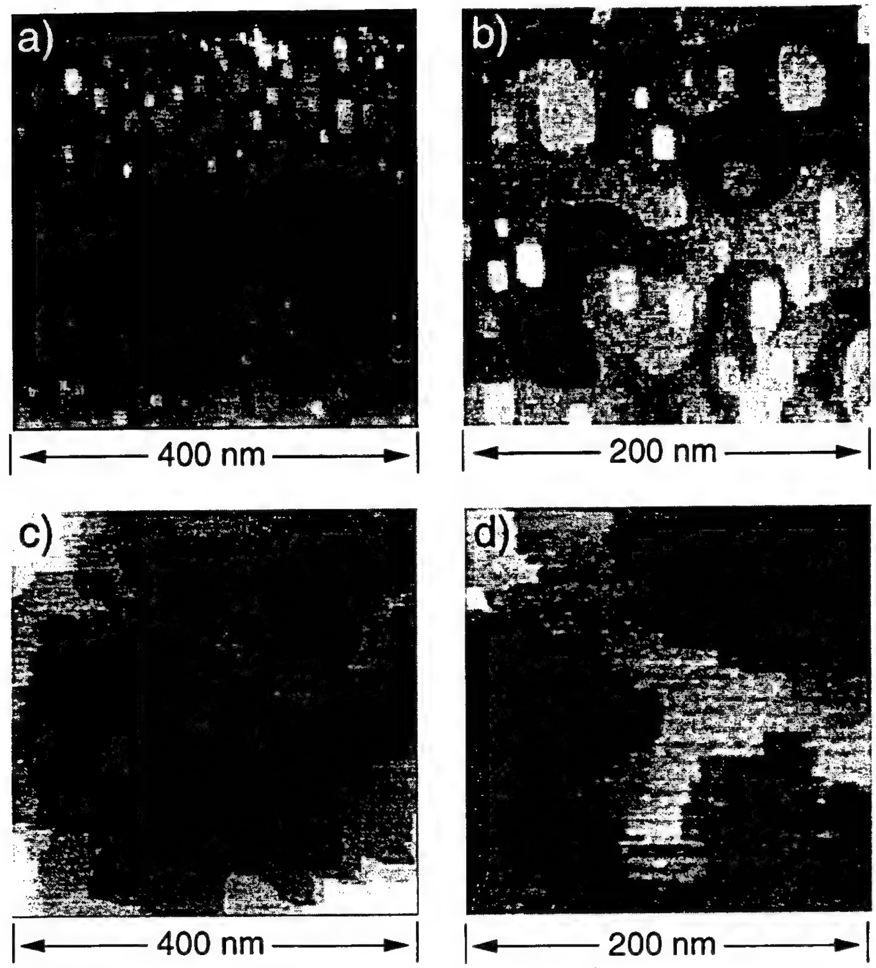


Fig. 4. Two STM images of an $Fe_{0.72}Al_{0.28}$ surface annealed to approximately 550°C, (a) and (b) and two of an $Fe_{0.74}Al_{0.24}$ surface annealed to approximately 650–700°C, (c) and (d). The sample from (a) and (b) exhibited a $(2\times2)$ diffraction pattern while (c) and (d) exhibited a $(5\times5)$ diffraction pattern. Both surfaces exhibited bilayer high steps, but the step edge kink density in (c) and (d) is much smaller than in (a) and (b). Notice also the crystalline anisotropy in (c) and (d).

ties of the sample could cause a change in the stray magnetic field produced by the current flow.

We conclude from the results shown in Fig. 5 of growth on a $(5\times5)$ surface that this surface has excess Fe on it and this excess Fe is incorporated into growth, changing the stoichiometry of the growth front enough to cause a monolayer growth mode, regardless of growth composition. Furthermore, if Fe incorporated only up to a saturation level around $x=0.75$, then any Fe not incorporated into the current layer would continue to ride the surface and be incorporated into the subsequent layers. This would cause monolayer oscillations to continue until the excess Fe was used up, at which point the growth mode would change back to a bilayer mode. The amount of growth needed before the growth would change back to a bilayer mode would then be dependent on the growth composition. For example, if growing $Fe_xAl_{1-x}$ at $x=0.5$, a monolayer of excess Fe would be incorporated after 4 ML of growth. If growing at higher Fe concentrations, such as at $x=0.62$, the excess Fe would be incorporated more slowly, over a period of 8 ML of growth. The

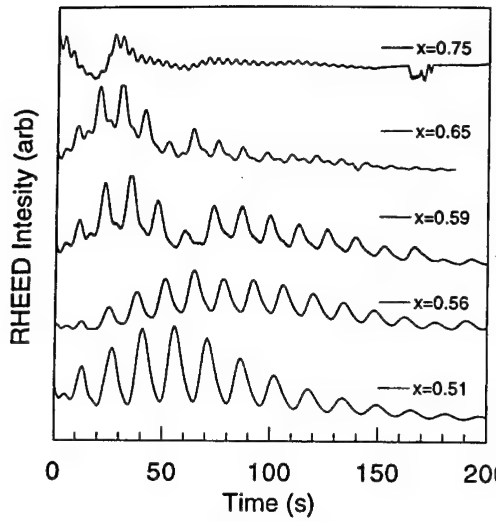


Fig. 5. RHEED oscillations due to growth on an annealed $Fe_xAl_{1-x}$ surface that exhibits a $(5\times5)$ diffraction pattern. It can be seen in that at a growth composition of $x=0.51$, the first oscillation is a monolayer oscillation and those after are bilayer oscillations. At higher Fe concentrations, the monolayer oscillations last longer until above $x=0.7$ where only monolayer oscillations are observed.

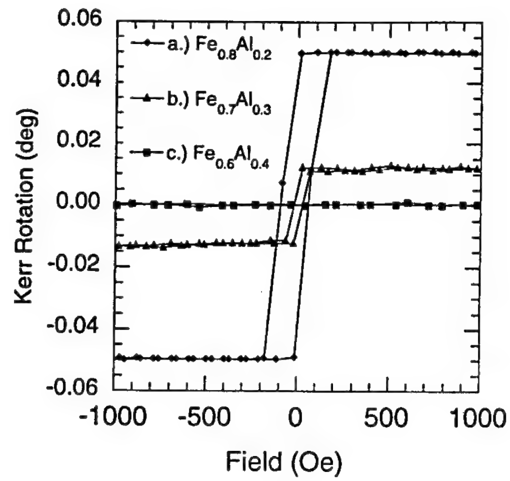


Fig. 6. Longitudinal Kerr loops of three samples of different compositions: (a) $x=0.8$, (b) $x=0.7$ and (c) $x=0.6$. The $x=0.6$ sample does not show Kerr rotation because it is non-magnetic. The other two samples are magnetic and show 100% in-plane remanence and a very quick magnetization reversal. The applied field at which the magnetization reverses is related to coercivity and it can be seen that the two samples have different coercivities.

incorporation time would increase exponentially as the growth composition approaches $x=0.75$, doubling every time the composition difference between $x=0.75$ is halved. This exponential effect appears to be the case in Fig. 5. From the measured transition times at these various coverages and assuming a saturation level of $x=0.75$, we estimate that a $(5\times5)$ surface has an excess of one half of a layer of Fe on it.

### 3.4. Magnetic properties

The magnetic properties of several thin films were examined to see if they possessed the same magnetic properties as bulk material and to see if they possessed magnetic properties that would be of interest for magnetic devices. Several samples spanning the composition range from ferromagnetic to non-magnetic were measured. Film thicknesses were 300 Å for the $x=0.6$ and $x=0.8$ sample and 500 Å for the $x=0.7$ sample. Fig. 6 shows the longitudinal Kerr measurements of these samples. The longitudinal Kerr rotation is a measure of the in-plane magnetization of the film. These measurements show that the $x=0.6$ sample was non-magnetic and the $x=0.7$ sample had a lower saturation magnetization than the $x=0.8$ sample, consistent with bulk properties. The $x=0.8$ sample showed a coercivity of about $H_c=75$ Oe and the $x=0.7$ sample showed a coercivity of about $H_c=12$ Oe. This data also indicates that the $x=0.7$ and $x=0.8$ sample have near 100% remanence at zero-applied field. Low field measurements of the $x=0.7$ confirmed this. From polar Kerr measurements, which measures out-of-plane magnetization, we found that the saturation field and maximum Kerr rotation were dependent on composition and no out-of-plane remanence was present. For the $x=0.8$ sample, the saturation field was $H_s=16$ KOe and maximum Kerr rotation was $\theta_k=0.228°$ whereas for the $x=0.7$ sample, $H_s=4.5$ KOe and $\theta_k=0.10°$. Thus, simply by varying $x$ in an expitaxial layer, one can vary the magnetic properties of the films. This could be useful to produce a giant magnetoresistance effect devices such as proposed by White in which a multilayer stack of high $H_c$

material followed by non-magnetic material and then a low $H_c$ material is needed [10].

## 4. Conclusions

We report on findings related to the nucleation, annealing, growth and magnetic properties of epitaxial $Fe_xAl_{1-x}$ on AlAs/GaAs. First, $Fe_xAl_{1-x}$ nucleates on an AlAs($3 \times 2$) surface in small clusters that are either disordered or randomly oriented. Second, it is necessary to grow a minimum thickness of $Fe_xAl_{1-x}$ on AlAs at low temperatures of about 200°C before annealing in order to obtain a surface in which RHEED oscillations can be seen with continued growth. Third, the growth mode of $Fe_xAl_{1-x}$ on an annealed $Fe_xAl_{1-x}$ surface is primarily determined by growth composition. Monolayer growth occurs above $x=0.7$ and bilayer growth occurs below that value. However, the initial growth behaviour also depends upon the surface reconstruction – a ($5 \times 5$) surface can force monolayer growth for at least several layers, probably due to an excess of Fe that slowly incorporates as subsequent layers are grown. Finally, samples with $x>0.7$ were shown to be ferromagnetic with 100% in-plane magnetization. $H_c$, $M_s$ and $\theta_k$ were found to be dependent on the Fe–Al ratio with all three increasing with increasing Fe concentration.

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# INVESTIGATION OF MBE GROWTH AND MAGNETIC PROPERTIES OF IRON ALUMINUM ON ALUMINUM ARSENIDE

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BRIAN EDWARD ISHAUG

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# 1. Abstract

Although several authors have reported previously on growth of FeAl on III-V semiconductors, the main purpose of our investigation was to apply scanning tunneling microscopy (STM) to the study of FeAl growth on III-V semiconductor. The idea was to use STM to better understand FeAl growth and help make sense out of the reflection high energy electron diffraction (RHEED) patterns that had been previously reported by Wowchak, Kuznia and Cohen for growth of FeAl on InP and FeAl on GaAs. These RHEED patterns where difficult, if not impossible, to interpret, particularly during initial nucleation where the RHEED pattern would disappear for a short period of time

Because STM is a relatively new technique, its capability to image an epitaxial metal surface was unknown. So, the first part of our investigation was to determine if STM could be used to provide useful information on the FeAl surface. We found that true atomic level resolution was almost impossible to obtain, but near atomic level resolution images could be obtained i.e. lateral resolution on the order of 10Å.. From these STM images it was possible to see the real space surface of FeAl and determine quantitative information of surface structure such as step heights, surface morphology and island sizes.

For our experiments, epitaxial growth was performed on an AlAs buffered GaAs substrate as opposed to an AlAs buffered InP substrate because GaAs is a much cheaper and easier substrate to deal with than InP. In addition, Kuznia et al. found that there was little difference between the growth dynamics of FeAl on GaAs and FeAl on InP, even though the lattice mismatches are quite different, +2.9% versus -0.9% respectively. On both substrates, the nucleation of FeAl on AlAs results in an incubation effect whereby the RHEED pattern disappears after a small amount of FeAl deposition, equivalent to about 1 bilayer of growth. With continued deposition, the RHEED pattern begins to reappear and after deposition equivalent to 2 to 3 bilayers of growth, the pattern intensity recovers to 30 to 50% of the original intensity.

We applied STM to study this initial nucletion stage, particularly at the point where no RHEED pattern is present. A STM image taken after deposition of FeAl

equivalent to 1 bilayer of growth, showed an AlAs surface covered with small clusters about 40Å in size and 3Å high. In comparison, an image taken after deposition of 3 bilayers, where a RHEED pattern is present, showed a rough, but apparently continuous surface with no visible indication of the AlAs surface beneath. This indicates that FeAl nucleates in small disordered clusters and a minimum of 2 to 3 bilayers of FeAl is needed in order for the FeAl film to become ordered and continuous.

Kuznia and Wowchak found that annealing films to 550°C improved the sharpness and brightness of the RHEED pattern which was interpreted as surface smoothening. This was confirmed with STM. A STM image of an unannealed surface revealed small islands and a large mean roughness. After annealing to 550°C, STM showed an atomically smooth film with terrace lengths in excess of 100Å and step heights of 3Å, which corresponds to a bilayer step.

In an effort to elucidate on the morphologic stability of FeAl on AlAs, several films were annealed to temperatures as high as 700°C. STM images of these surfaces again showed an atomically smooth surface with no evidence of pin holes. This result is important for applications where high temperature morphologic stability is important such as overgrowth of semiconductor. It is worth noting that in higher Fe composition films, 70-80% Fe, the RHEED pattern changed from a $(2\times2)$ reconstruction to a $(5\times5)$ reconstruction after annealing to 700°C. This is thought due to Fe segregating to the surface which changes the surface reconstruction. Unfortunately, the resolution of our STM images are not high enough to see the surface reconstruction.

It is known that FeAl growth on FeAl occurs in 2 distinct modes, monolayer by monolayer and bilayer by bilayer, depending on compositoin. We attempted STM on several surface which exhibited monolayer and several which exhibited bilayer oscillations. Unfortunately, our results were inconclusive as the surface after growth is too rough to get good STM images. After annealing the films, the surface became smooth enough to get good images, but only bilayer high steps were observed We did, however, eluxidate on the conditions in which monolayer and bilayer oscillations are observed. In addition to growth composition, it is thought that excess Fe on the surface

also affects the growth mode. The theory is when growing FeAl on an Fe rich FeAl surface, the oscillations start in a monolayer mode and after a several layers of growth, change to bilayer mode because the excess Fe slowly incorporates into the growth front, changing it enough to cause monolayer oscillations. This incorporation of Fe is limited to some maximum composition of Fe, perhaps x=0.75, and any Fe not used up continues to rides the surface. Once the excess Fe is used up, the growth changes back to bilayer mode.

We also report the results of magneto-optic Kerr effect measurements performed by R.F.C Farrow et al. at IBM Almaden Research Center on three of our FeAl film with a composition of 60%, 70% and 80% Fe and thickness greater than 300Å. The general magnetic properties were found to be consistent with bulk magnetic properties. From longitudinal and polar Kerr hysteresis loops, we found that the 60% Fe film was non-magnetic. The other two films were magnetic and had 100% in-plane remnant magnetization, but no out of plane remnant magnetization. This is expected due to the shape anisotropy. The maximum Kerr rotation ( $\theta_{k\,max}$ ), saturation field ($H_s$) and in-plane coercivity ($H_c$) where found to be dependent on composition with the 80% Fe sample exhibiting a larger $\theta_{k\,max}$ , $H_s$ and $H_c$ than the 70% Fe sample. From Kerr spectra, we found some evidence of magnetic ordering in the 80% Fe sample.